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# (54) HERBICIDAL COMPOSITIONS AND METHOD OF USING THE SAME

HERBIZIDE ZUSAMMENSETZUNGEN UND ANWENDUNGSVERFAHREN COMPOSITIONS HERBICIDES ET MODE D'UTILISATION

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- (56) References cited:

EP-A1- 0 595 126 EP-A1- 0 862 857 EP-A2- 0 325 054 JP-A- 7 242 510 JP-A- 60 075 324 US-A- 4 080 191

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### Description

[0001] This invention relates to a herbicidal composition having a rapid action, an improved effect and a long-term stability of preparation.

[0002] Among the light-inducing herbicidal compositions represented by the general formula (I), the 3-substituted phenylpyrazole derivatives represented by the general formula (I-1) are known compounds disclosed in JP-A-3-163063 and JP-A-4-211065, wherein it is mentioned that these compounds have, as herbicides for foliage treatment, an outstanding herbicidal activity on the general broad-leaved weeds noxious in the upland farming.

[0003] It is known that a similar herbicidal activity is exhibited by a number of compounds resembling the general formula (I-1) in the over-all chemical structure and relative configuration of substituents used as light-inducing or protox-inhibiting herbicides [Anderson et al., ACS Symposium Series, Vol. 559, Porphric Pesticides, S.O. Duke and C.A. Robeiz eds., p. 18-34 (1994)]. It is also known that a resistance to these light-inducing herbicides can generally be given to plants by introducing a specific gene into the plants (International Patent Application WO98/29554).

[0004] On the other hand, organophosphorus herbicidal compounds such as the N-(phosphonomethyl)-glycine or salts thereof disclosed in JP-A-47-39538 and JP-A-57-95994, the 4-[hydroxy(methyl)phosphino]-L-homoalanine or salts thereof disclosed in JP-A-57-26564 and the 4-[hydroxy(methyl)phosphino]-L-homoalany-L-alanyl-L-alanine or salts thereof disclosed in JP-A-50-23282, etc. are well known as non-selective herbicides for foliage treatment.

[0005] Further, JP-A-7-242510 discloses herbicidal compositions containing a 3-substituted phenylpyrazole derivative and an organophosphorus herbicidal compound.

**[0006]** Further, Weed Sci. Soc. 25, (1977), p.275-287 refers to the adjuvant effect of alcohol alkoxylates on N-(phosphonomethyl)glycine. However, this technique is insufficient in the adjuvant effect and the alcohol alkoxylates are not readily compatible with aqueous solution of N-(phosphonomethyl)glycine.

[0007] EP-A-0 595 126 discloses a herbicidal composition containing as the active ingredient at least one organophosphorus compound and 3-substituted phenylpyrazole derivative.

[0008] EP-A-0 862 857 relates to compositions of nonionic siloxane alkoxylates with aminopolyalkyleneoxide surfactants which are useful in treating plants, for instance as adjuvants for pesticides including in particular herbicides.

**[0009]** EP-A-0 325 054 discloses a nonionic composition which can be used to prepare a wide range of emulsions or dispersion of solids in liquids, e.g. it can be used to emulsify or disperse agrochemicals such as herbicides. The composition is a reaction product of at least four different compounds including a poly(oxyalkylene), a compound which contains at least two reactive hydrogen atoms, a carboxylic acid (derivative) having at least two carboxylic groups and a compound selected from a mono-carboxylic acid, a mono-hydroxy compound and mono-amino compound.

**[0010]** JP 60-075324 describes a preparation containing as a primary component an alkylene oxide adduct of a compound having two amino groups in the molecule which is used for aqueous suspension type preparations for agricultural chemicals such as insecticides, disinfectants and herbicides.

[0011] US 4,080,191 relates to a pesticidal formulation comprising a pesticidal agent, an anionic or non-ionic surfactant and a water-immiscible solvent/diluent The formulation is useful in combating pests on or directly beneath water.

[0012] Thus, it has been demanded to improve the herbicidal effect and rapid action of a herbicidal composition containing a light-inducing herbicidal compound (especially a 3-substituted phenylpyrazine derivative) and an organ-ophosphorus herbicidal compound.

[0013] The present inventors have conducted extensive studies with the aim of solving the problems mentioned above to find that, by adding an ethylenediamine alokoxylate and an alcohol alkoxylate as surfactants to a herbicidal composition containing one or more compounds selected from light-inducing herbicidal compounds, especially one or more compounds selected from the 3-substituted phenylpyrazole derivatives represented by the general formula (I-1), and the organophosphorus herbicidal compounds as active ingredients, the rapid action of the composition can be made excellent and the effect of the composition can be improved by the synergistic action of the active ingredients and the surfactants, besides there can be obtained a herbicidal composition keeping stable for a long period of time. Based on this finding, this invention has been accomplished.

**[0014]** The present invention relates to a herbicidal composition containing one or more compounds selected from light-inducing herbicidal compounds and one more compounds selected from organic herbicidal compounds as active ingredients: wherein said light-inducing herbicidal compound is a compound represented by the following general formula (I):

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wherein P represents any one of P1 to P9:

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$$CI \longrightarrow X^1$$
  $CI \longrightarrow X^1$   $CI \longrightarrow X^1$   $COOR^5$   $P^2$   $P^3$   $P^4$ 

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$$CH_3$$
  $COOR^6$   $P^5$   $P^6$   $CH_3$   $COOR^6$   $P^7$ 

$$CI$$
 $X^1$ 
 $CI$ 
 $X^1$ 
 $CI$ 
 $X^1$ 
 $COOCH(R^4)CO-Y^1R^5$ 
 $P^8$ 
 $P^9$ 

wherein  $X^1$ , Y,  $Y^1$ ,  $Y^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  are as defined later, and Q represents any one of  $Q^1$  to  $Q^{11}$ :

wherein

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R<sup>1</sup> represents a (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

R<sup>2</sup> represents a hydrogen atom, a (C<sub>1</sub>-C<sub>6</sub>) alkyl group or a halo (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

 $R^3$  represents a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group, a  $(C_3-C_6)$  cycloalkyl group, a  $(C_2-C_6)$  alkenyl group or a  $(C_2-C_6)$  alkynyl group;

R<sup>4</sup> represents a hydrogen atom or a (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

R<sup>5</sup> represents a hydrogen atom, a (C<sub>1</sub>-C<sub>6</sub>) alkyl group, a halo (C<sub>1</sub>-C<sub>6</sub>)

alkyl group, a (C2-C6) alkenyl group or a (C2-C6) alkynyl group;

R<sup>6</sup> represents a (C<sub>1</sub>-C<sub>6</sub>) alkyl group, a (C<sub>2</sub>-C<sub>6</sub>) alkenyl group or a (C<sub>2</sub>-C<sub>6</sub>) alkynyl group;

X<sup>1</sup> and X<sup>2</sup> represent halogen atoms which may be the same or different;

Y represents -O-, -S-, -SO- or -SO<sub>2</sub>-;

Y1 represents -O- or -S-;

Y2 represents -O-, -S- or -NH-;

and n represents an integer of 0 to 1

and wherein said organophosphorus herbicidal compound is N-(phosphono-methyl)glycine or a salt thereof, 4-[hydroxy (methyl)phosphino]-L-homoalanine or a salt thereof, or 4-[hydroxy(methyl)phosphino]-L-homoalanyl-L-alanyl-L-alanine or a salt thereof,

characterized in that the herbicidal composition further contains an ethylenediamine alkoxylate and an alcohol alkoxylate as surfactants wherein said alcohol alkoxylate is a mixture of one or more compounds selected from the group consisting of primary alcohol ethoxylate, primary alcohol propoxylate, primary alcohol ethoxylate propoxylate, secondary alcohol ethoxylate, secondary alcohol ethoxylate, tertiary alcohol ethoxylate, tertiary alcohol ethoxylate, tertiary alcohol ethoxylate.

[0015] Particularly, the present invention relates to a herbicidal composition containing, as the light-inducing herbicidal compound, one or more compounds selected from the group consisting of the 3-substituted phenylpyrazole derivatives represented by the following general formula (I-1):

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$$X_1$$
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_2$ 
 $X_3$ 
 $X_4$ 
 $X_2$ 
 $X_2$ 
 $X_3$ 
 $X_4$ 
 $X_4$ 
 $X_5$ 
 $X_5$ 
 $X_7$ 
 $X_8$ 
 $X_$ 

wherein R represents -Y¹-R³ (wherein R³ represents a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkynyl group; and Y¹ represents -O- or -S-), -Y²-CH(R⁴) CO-OR⁵ (wherein R⁴ represents a hydrogen atom or a  $(C_1-C_6)$  alkyl group; R⁵ represents a hydrogen atom, a  $(C_1-C_6)$  alkyl group, halo  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkynyl group; and Y² represents -O-, -S- or -NH-), -COOCH (R⁴) CO-Y¹-R⁵ (wherein R⁴, R⁵ and Y¹ are as defined above), or -COOR⁶ (wherein R⁶ represents a  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkynyl group); R¹ represents a  $(C_1-C_6)$  alkyl group; R² represents a hydrogen atom, a  $(C_1-C_6)$  alkyl group or a halo  $(C_1-C_6)$  alkyl group; X¹ and X² represent halogen atoms which may be the same or different; Y represents -O-, -S-, -SO- or -SO₂-; and n represents an integer of 0 to 1.

**[0016]** Among the substituents in the general formula (I) and general formula (I-1), the term " $(C_1-C_6)$  alkyl group" means a straight or branched chain alkyl, group having 1-6 carbon atoms such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, n-pentyl or n-hexyl "halo  $(C_1-C_6)$  alkyl group" means a straight or branched chain alkyl group having 1-6 carbon atoms substituted with one or more halogen atoms which are selected from the group consisting of chlorine atom, fluorine atom, iodine atom and bromine atom and may be the same or different; " $(C_2-C_5)$  alkenyl group" means a straight or branched chain alkenyl group having 2-6 carbon atoms; and " $(C_2-C_6)$  alkynyl group" means a straight or branched chain alkynyl group having 2-6 carbon atoms.

[0017] As examples of the light-inducing herbicidal compounds of this invention, the following ones can be referred to:

- (1) 3-(4-Chloro-5-(cyclopentyloxy)-2-fluoro-phenyl)-5-isopropylidene-1,3-oxazolidindione (general name: pentox-azone)
- (2) Pentyl [2-chloro-5-(cyclohexa-1-ene-1,2-dicarboximide)-4-fluorophenoxy]-acetate (general name: flumicloracpentyl)
- (3) 7-Fluoro-6-[(3, 4, 5, 6-tetrahydro) phthalimido]-4-(2-propynyl)-1,4-benzoxazin-3(2H)-one (general name: flumioxazin)
- (4) Ethyl (RS)-2-chloro-3-[2-chloro-5-(4-fluoromethyl-4,5-dihydro-3-methyl-5-oxo-1H-1,2,4-triazol-1-yl)-4-fluorophenyl]propionate (general name: carfentrazone-ethyl)
- (5) 2',4'-Dichloro-5'-(4-difluoromethyl-4,5-dihydro-3-methyl-5-oxo-1H-1,2,4-triazol-1-yl)-methane-sulfonanilide (general name: sulfentrazone)
- (6) 2-(2,4-Dichloro-5-propyn-2-yloxyphenyl)-5,6,7,8-tetrahydro-1,2,4-triazolo[4,3-a]pyridin-3(2H)-one (general name: azafenidin)
- (7) 5-tert-Butyl-3-[2,4-dichloro-5-(propyn-2-yloxy)phenyl]-1,3,4-oxadiazole (general name: oxadiargyl)
- (8) 5-tert-Butyl-3-(2,4-dichloro-5-isopropoxy-phenyl)-1,3,4-oxadiazol-2(3H)-one (general name: oxadiazon)
- (9) Methyl [2-chloro-4-fluoro-5-(5,6,7,8-tetrahydro-3-oxo-1H,3H-[1,3,4]thiadiazolo[3,4-a]pyriadzin-1-ylideneamino)phenylthio]acetate (general name: fluthacet-methyl) etc.

[0018] As the compound groups exhibiting a similar 5 activity, the following diphenyl ether type compounds are known:

nitrofen (general name), bifenox (general name), oxyfluorfen (general name), acifluorfen (general name), fomesafen (general name), etc. Besides above, the following 3-substituted phenylpyrazole derivatives represented by the general formula (I-1) can also be referred to:

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General Formula (I-1)

[0019]

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10 (I-1)

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Table 1

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(R <sub>1</sub> = CH <sub>3</sub> )							
No.	R	R <sub>2</sub>	X <sub>1</sub>	X <sub>2</sub>	(Y) n	Property	
1	OCH <sub>2</sub> CH=CH <sub>2</sub>	CH <sub>3</sub>	CI	CI	s	nD 1.6131(25.3°C)	
2	OCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5536(28.4°C)	
3	OCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 63.7-64.1°C	
4	SCH <sub>2</sub> CH=CH <sub>2</sub>	CH <sub>3</sub>	CI	CI	s	Paste	
5	SCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 52.0-55.0°C	
6	SCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5670 (17.9°C)	
7	OCH <sub>2</sub> C≡CH	CH <sub>3</sub>	CI	CI	s	m.p. 71.5°C	
8	OCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	0	m.p. 84.0°C	
9	OCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	F	CI	0	m.p. 98.0-98.1°C	
10	SCH <sub>2</sub> C≡CH	CH <sub>3</sub>	CI	CI	s	m.p. 94.5°C	
11	SCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	0	m.p. 127-129°C	
12	SCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	F	CI	0	m.p. 82.8°C	
13	OCH₂COOCH3	CH <sub>3</sub>	CI	CI	s	m.p. 126.2°C	
14	OCH <sub>2</sub> COOCH <sub>3</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 119.8°C	
15	OCH <sub>2</sub> COOCH <sub>3</sub>	CHF <sub>2</sub>	CI	Br	0	m.p. 133.8°C	
16	OCH₂COOCH3	CHF <sub>2</sub>	F	CI	0	m.p. 122.8-123.1°C	
17	OCH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>	CI	CI	s	m.p. 106.5°C	
18	OCH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 102.3°C	
19	OCH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 127.6°C	
20	OCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-n</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 89.7°C	
21	OCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-n</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 97.6-97.8°C	
22	OCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 106.0°C	
23	OCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 120.3-120.5°C	
24	OCH <sub>2</sub> COOCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 84.7°C	
25	OCH <sub>2</sub> COOCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 89.2-89.4°C	
26	OCH <sub>2</sub> COOCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	0	m.p. 119.6°C	
27	OCH <sub>2</sub> COOCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	F	CI	0	m.p. 99.0°C	
28	осн(сн₃)соон	CH <sub>3</sub>	CI	CI	s	m.p. 191-194°C	
29	OCH(CH3) COOCH3	CH <sub>3</sub>	CI	CI	s	m.p. 90-93°C	
30	OCH(CH3)COOCH3	CHF <sub>2</sub>	F	CI	0	m.p. 95.6°C	
31	OCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>	CI	CI	s	nD 1.5763 (28.8°C)	
32	OCH(CH <sub>3</sub> )COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5238(25.7°C)	
33	OCH(CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	Br	0	nD 1.5396 (20.8°C)	
34	OCH(CH <sub>3</sub> )COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	m.p. 67.0-67.2°C	

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Table 1 (continued)

	$(R_1 = CH_3)$						
	No.	R	R <sub>2</sub>	X <sub>1</sub>	X <sub>2</sub>	(Y) n	Property
5	35	OCH (CH <sub>3</sub> ) COOC <sub>3</sub> H <sub>7-i</sub>	CH <sub>3</sub>	CI	CI	S	m.p. 87-90°C
	36	SCH (CH₃) COOCH₃	CHF <sub>2</sub>	CI	CI	0	nD 1. 5654 (19.8°C)
	37	SCH (CH <sub>3</sub> ) "COCCH <sub>3</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5494 (25.0°C)
	38	SCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5565 (28.0°C)
10	39	SCH(CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5328 (18.0°C)
	40	NHCH(CH <sub>3</sub> ) COOCH <sub>3</sub>	CH <sub>3</sub>	CI	CI	S	m.p. 144.2°C
	41	NHCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>	CI	CI	S	Paste
	42	NHCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5371 (23.4°C)
	43	NHCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1. 5264 (26.6°C)
15	44	COOCH₂COOCH₃	CHF <sub>2</sub>	CI	CI	0	m.p. 74.4°C
	45	COOCH₂COOCH₃	CHF <sub>2</sub>	F	CI	0	nD 1.5350(27.3°C)
	46	COOCH₂COSCH₃	CHF <sub>2</sub>	CI	CI	0	-
	47	COOCH₂COSCH₃	CHF <sub>2</sub>	F	CI	0	
20	48	COOCH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 57.2°C
	49	COOCH <sub>2</sub> COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5362 (23.4°C)
	50	COOCH <sub>2</sub> COSC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5763 (20.7°C)
	51	COOCH <sub>2</sub> COSC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5536(27.3°C)
	52	COOCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1. 5289 (24.0°C)
25	53	COOCH <sub>2</sub> COOC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	F	CI	0	
	54	COOCH <sub>2</sub> COSC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5684 (20.2°C)
	55	COOCH <sub>2</sub> COSC <sub>3</sub> H <sub>7-i</sub>	CHF <sub>2</sub>	F	CI	0	
	56	COOCH <sub>2</sub> COOCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 45.4°C
30	57	COOCH <sub>2</sub> COOCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	F	CI	0	
	58	COOCH <sub>2</sub> COOCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	0	m.p. 79.3°C
	59	COOCH <sub>2</sub> COOCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	F	CI	0	
	60	COOCH (CH <sub>3</sub> ) COOCH <sub>3</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1. 5370 (25.7°C)
	61	COOCH (CH <sub>3</sub> ) COOCH <sub>3</sub>	CHF <sub>2</sub>	F	CI	0	nD 1. 5314 (23.0°C)
35	62	COOCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1. 5672 (26.0°C)
	63	COOCH (CH <sub>3</sub> ) COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5212 (14.1°C)
	64	COOCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	0	m.p. 78.5°C
	65	COOCH <sub>3</sub>	CHF <sub>2</sub>	CI	CI	0	m.p. 63.9°C
40	66	COOCH <sub>3</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5430(17.0°C)
	67	COOC <sub>2</sub> H <sub>5</sub>	CH <sub>3</sub>	CI	CI	S	nD 1.6029(20.1°C)
	68	COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	CI	CI	0	nD 1.5446 (26.8°C)
	69	COOC <sub>2</sub> H <sub>5</sub>	CHF <sub>2</sub>	F	CI	0	nD 1.5320 (21.0°C)
	70	OCH <sub>2</sub> CH=CH <sub>2</sub>	CHF <sub>2</sub>	CI	CI	NH	m.p. 80.6°C
45	71	OCH <sub>2</sub> C≡CH	CHF <sub>2</sub>	CI	CI	NH	m.p. 118.9°C
	72	OCH <sub>2</sub> COOCH <sub>3</sub>	i-C <sub>3</sub> H <sub>7</sub>	CI	CI	-	Paste
	73	OCH <sub>2</sub> CH=CH <sub>2</sub>	i-C <sub>3</sub> H <sub>7</sub>	CI	CI	-	Paste
	74	OCH <sub>2</sub> C≡CH	i-C <sub>3</sub> H <sub>7</sub> ,	CI	CI	-	Paste
50	75 70	SCH <sub>2</sub> COOCH <sub>3</sub>	t-C <sub>4</sub> H <sub>9</sub>	CI	CI	-	Paste
1	76	OCH <sub>2</sub> CH=CH <sub>2</sub>	CH <sub>2</sub> Br	CI	CI	-	Paste

[0020] Among the 3-substituted phenylpyrazole derivatives of the present invention represented by the general formula (I-1), preferred is the compound No. 19.

[0022] N-(phosphonomethyl)glycine or salts thereof, such as N-(phosphonomethyl)glycine isopropylamine salt (hereinafter, referred to as "Compound A"), or ammonium salt thereof or N-(phosphonomethyl)glycine trimethylsulfo-

<sup>[0021]</sup> On the other hand, as the organophosphorus herbicidal compounds used in the present invention, the following can be referred to:

nium salt (hereinafter, referred to as "Compound B");

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[0023] 4-[hydroxy(methyl)phosphino]-DL-homoalanine or salts thereof such as 4-[hydroxy(methyl)phosphino]-DL-homoalanine ammonium salt (hereinafter, referred to as "Compound C");

[0024] 4-[hydroxy(methyl)phosphino]-L-homoalanyl-L-alanyl-L-alanine or salts thereof, such as 4-[hydroxy-(methyl) phosphino]-L-homoalanyl-L-alanyl-L-alanine sodium salt (hereinafter, referred to as "Compound D"); etc.

[0025] Among these organophosphorus herbicidal compounds, preferred is N-(phosphonomethyl)glycine isopropylamine salt.

[0026] As the ethylenediamine alkoxylates used as a surfactant in this invention, the compounds represented by the following general formula (II):

 $H - (Po)_a - (Eo)_b$   $N - CH_2 - CH_2 - N$   $(Eo)_f - (Po)_g - H$   $H - (Po)_d - (Eo)_e$   $(Eo)_h - (Po)_i - H$  (II)

wherein Eo represents  $-O-CH_2CH_2-$ , Po represents  $-O-CH(CH_3)CH_2-$ , and a, b, d, e, f, g, h, and i represent integers of 1 to 20 which may be the same or different, can be referred to. As preferable examples thereof, Teric 170 (manufactured by Huntsman Co.) and Teric 173 (manufactured by the same company as above) can be referred to. One or more species of the above-mentioned ethylenediamine alkoxylates may be selected and put to use.

[0027] Moreover, as the ethylendediamine alkoxylates can be further used an adduct of ethylenediamine and polyoxyethylene, and adduct of ethylenediamine and polyoxypropylene or an adduct of ethylenediamine and a polyoxyethylene-polyoxypropylene copolymer. examples of the alcohol alkoxylates include the following: Noigen ET-165 (manufactured by Daiichi Kogyo Seiyaku K.K.), Adekatol SO-135 (manufactured by Asahi Denka K.K.), Noigen ET-115 (manufactured by Daiichi Kogyo Seiyaku K.K.), NK (Newkalgen)-D-1107S (manufactured by Takemoto Yushi K.K.), Lutensol T08 (manufactured by BASF Co.), TO-347 (manufactured by Nippon Nyuukazai K.K.), etc. One or more species of such alcohol alkoxylates may be selected and put to use.

[0028] In the herbicidal composition of the present invention, compounding ratios of the ingredients are as follows. That is, in 100 parts by weight of the herbicidal composition, the light-inducing herbicidal compound is used in an amount of 0.01-10.0 parts by weight and preferably 0.1-2.0 parts by weight; the organophosphorus herbicidal compound is used in an amount of 1.0-60.0 parts by weight and preferably 5.0-40.0 parts by weight; the ethylenediamine alkoxylate is used in an amount of 0.1-25 parts by weight and preferably 10-25 parts by weight; and the alcohol alkoxylate is used in an amount of 0.1-15.0 parts by weight and preferably 0.1-5.0 parts by weight.

**[0029]** When the herbicidal composition of the present invention is put to use, the composition is made into an appropriate preparation form in accordance with the purpose according to the conventional method in the pesticide preparation, and then put to use. For instance, the composition is blended with a solid carrier, a liquid carrier or other necessary adjuvants, and the blended mixture thus obtained is made into a preparation form such as a suspension concentrate in which the active ingredient exists in the state of suspended fine particles, or a wettable powder, or a water dispersible granule, or an aqueous preparation and then put to use.

[0030] Further, it is also possible, if desired, to blend the composition of the present invention with an emulsifier comprising the same surfactant as used in the present invention in an amount of 0.1-99.0 parts by weight and preferably 25.0-75.0 parts by weight per part by weight of the light-inducing herbicide compound of general formula (I), and a commercially available preparation containing an organophosphorus compound as an active ingredient thereof, at the time of preparing a liquid preparation to be sprayed, and thereafter to put to use.

[0031] When using a suspension concentrate or a water dispersible granule which contains, as the active ingredients thereof, a finely pulverized material comprising 0.01-10 parts by weight of the light-inducing herbicidal compound represented by the general formula (I) without surfactant, and 1-50 parts by weight of an organophosphorus herbicidal compound; it is also possible, if desired, to make the same surfactant as used in this invention into a solution elsewhere and to mix the resulting solution with the suspension concentrate or water dispersible granule of this invention at the time of preparing a liquid mixture to be sprayed, at a ratio of 10-90 parts by weight of the surfactant solution per part by weight of the light-inducing herbicidal compound represented by the general formula (I).

#### **EXAMPLES**

[0032] Next, typical examples and test examples of the present invention will be presented below. This invention is

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by no means limited by these examples.

[0033] As used in the examples, the term "part" and "parts" are by weight.

(Base)

[0034]

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Compound No.19	40.0 parts
Neocol YSK (manufactured by	
Daiichi Kogyo Seiyaku)	1.0 part
Sorpol 7425 (Manufactured by	
Toho Kagaku Kogyo)	3.0 parts
Propylene glycol	3.0 parts
Silicone KM-73 (manufactured by	
Shin'etsu Kagaku)	0.5 part
Proxel GXL (manufactured by	
Zeneca Japan)	0.1 part
Rhodopol 23 (manufactured by	
Rhodia Nikka)	0.02 part
Water	Balance
Total	100 parts

[0035] Each of the mixtures having the above-mentioned compounding ratios was finely pulverized by means of Dyno-Mill (manufactured by Bachofen AG) filled with 0.3 mm ceramic beads (Toreceram, manufactured by Toray K. K.) to prepare a suspension composition containing 40% of Compound No.19 as fine particles having a mean particle diameter of 0.3 μm.

[0036] Using the suspension concentrate thus obtained as a base, the compositions of the following Examples 1-12 were prepared.

## Example 1

## [0037]

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	Base of Compound No.19 (40.0%)	0.41 part
	Compound A (62.0% aqueous solution)	50.80 parts
	Teric 170	10.00 parts
40	Noigen ET-165	2.00 parts
40	Propylene glycol	2.50 parts
	Soprophor DSS/7-60 (dispersant;	
	manufactured by Rhodia Nikka Co.)	0.83 part
	N-Methylpyrrolidone (solvent)	0.15 part
45	Silicone KM-73 (antifoaming agent)	0.50 part
	Proxel GXL (preservative)	0.10 part
	Attagel 50 (thickener; manufactured	
	by Neolite Kosan)	10.0 parts
	Rhodopol 23 (thickener)	0.10 part
50	Water	Balance
	Total	100.00 parts

[0038] A mixture of the above-mentioned formulation was thoroughly homogenized by means of a mixer such as
Autohomomixer (manufactured by Tokushu Kika K.K.) or the like to obtain a suspension concentrate containing 0.15% of Compound No.19 and 30% of Compound A.

## Examples 2-12

**[0039]** Suspension concentrate were prepared by repeating Example 1, except that the ethylenediamine alkoxylate, alcohol alkoxylate and organophosphorus herbicidal compound used in Example 1 were replaced with those shown in Table 2.

Table 2

	Table 2	
Example No.	Organophosphorus herbicidal compound Ethylenediamine alkoxylate Alcohol alkoxylate	Compounding ratio (parts by wt.)
2	Compound A	30.00
	Teric 170	10.00
	Adekatol SO-135 (HLB=13)	2.00
3	Compound A	30.00
	Teric 170	10.00
	Noigen ET-115 (HLB=11)	2.00
4	Compound A	30.00
	Teric 170	10.00
	NK (Newkalgen) D-1107S	2.00
5	Compound A	30.00
	Teric 170	10.00
	Lutensol TO8 (HLB=13)	2.00
6	Compound A	30.00
	Teric 170	10.00
	TO-347	2.00
7	Compound A	30.0
	Teric 173	10.00
	Noigen ET-165 (HLB=216)	2.00
8	Compound A	30.0
	Teric 173	10.00
	Adekatol SO-135	2.00
9	Compound A	30.0
	Teric 173	10.00
	Noigen ET-115	2.00
10	Compound B	30.0
	Teric 173	10.00
	NK-D-1107S	2.00
11	Compound C	30.0
	Teric 173	10.00
	Lutensol TO8	2.00
12	Compound D	30.0
	Teric 173	10.00
	TO-347	2.00

Comparative Example 1

[0040] A commercial preparation containing Compound A as an active ingredient (glyphosate isopropylamine salt solution) was used.

### Comparative Example 2

[0041] A composition was prepared by repeating Example 1, except that the ethylenediamine alkoxylate was not used and the amount of the alcohol alkoxylate was altered from 2 parts to 12 parts.

### Comparative Example 3

[0042] A composition was prepared by repeating Example 1, except that the alcohol alkoxylate was not used and the amount of the ethylenediamine alkoxylate was altered from 10 parts to 12 parts.

#### Test Example 1

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[0043] Barnyard grass (Echinochloa crus-galli) and cocklebur (Xanthium strumarium) were cultured in a plastic-made pot having a diameter of 12 cm. When the plants had reached 7-leaved stage or a later stage, a test agent solution adjusted to a prescribed concentration was sprayed, and the herbicidal effect was visually evaluated by the naked eye on the third day and twenty first day after the treatment (0: no herbicidal effect; 100: withering).

[0044] The results are summarized in Table 3.

Table 3

20	Example	C	ompound (g/ha)	3 days after		21 days after	
	No.	19	Phospho-rous type	Barnyard grass	Cocklebur	Barnyard grass	Cocklebur
	1	5	1000	68	85	100	100
	2	5	1000	65	85	100	100
25	3	5	1000	60	85	100	100
	4	5	1000	65	85	100	100
	5	5	1000	68	83	100	100
	6	5	1000	63	85	100	100
30	7	5	1000	65	85	100	100
	8	5	1000	68	85	100	100
	9	5	1000	68	83	100	100
	10	5	1000	65	80	100	100
	11	5	1000	68	83	100	100
35	12	5	1000	65	85	100	100
	Comparative	0	1000	20	20	85	100
	Example 1						
	Comparative	5	1000	65	85	90	85
40	Example 2						
	Comparative	5	1000	20	40	80	85
	Example 3						

[0045] It has been found that, in a composition comprising a mixture of light-inducing herbicidal compound, especially a 3-substituted phenylpyrazole derivative, and an organophosphorus herbicidal compound, an excellent rapid herbicidal action and an improvement of the herbicidal effect can be achieved by incorporating an ethylenediamine alkoxylate and an alcohol alkoxylate into the composition.

# 50 Claims

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 A herbicidal composition containing one or more compounds selected from light-inducing herbicidal compounds and one or more compounds selected from organophosphorus herbicidal compounds as active ingredients thereof, wherein said light-inducing herbicidal compound is a compound represented by the following general formula (I):

P-Q (I)

wherein P represents any one of P1 to P9:

$$CI \longrightarrow \begin{array}{c} X^1 \\ CI \longrightarrow \\ R^4 \\ P^2 \end{array}$$

$$COOR^5 \qquad P^2$$

$$R^6 \qquad P^3 \qquad R^6 \qquad P^4$$

wherein  $X^1$ , Y,  $Y^1$ ,  $Y^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  are as defined later, and Q represents any one of  $Q^1$  to  $Q^{11}$ :

wherein

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R1 represents a (C1-C6) alkyl group;

 $R^2$  represents a hydrogen atom, a  $(C_1-C_6)$  alkyl group or a halo  $(C_1-C_6)$  alkyl group;

 $R^3$  represents a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group, a  $(C_3-C_6)$  cycloalkyl group, a  $(C_2-C_6)$  alkenyl group or a  $(C_2-C_6)$  alkynyl group;

R<sup>4</sup> represents a hydrogen atom or a (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

 $R^5$  represents a hydrogen atom, a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkynyl group; or a  $(C_2-C_6)$  alkynyl group;

R<sup>6</sup> represents a (C<sub>1</sub>-C<sub>6</sub>) alkyl group, a (C<sub>2</sub>-C<sub>6</sub>) alkenyl group or a (C<sub>2</sub>-C<sub>6</sub>) alkynyl group;

X<sup>1</sup> and X<sup>2</sup> represent halogen atoms which may be the same or different;

Y represents -O-, -S-, -SO- or -SO2;

Y<sup>1</sup> represents -O- or -S-;

Y2 represents -Q-, -S- or -NH-;

and n represents an integer of 0 to 1,

and wherein said organophosphorus herbicidal compound is N-(phasphono-methyl)glycine or a salt thereof, 4-[hydroxy(methyl)phosphino]-L-homoalanine or a salt thereof, or 4-hydroxy(memyl)phosphino]-L-homoalanyl-L-alanyl-L-alanine or a salt thereof,

characterized in that the herbicidal composition further contains an ethylenediamine alkoxylate and an alcohol alkoxylate as surfactants

wherein said alcohol alkoxylate is a mixture of one or more compounds selected from the group consisting of primary alcohol ethoxylate, primary alcohol propoxylate, primary alcohol ethoxylate propoxylate, secondary alcohol ethoxylate propoxylate, tertiary alcohol ethoxylate, tertiary alcohol propoxylate and tertiary alcohol ethoxylate propoxylate.

2. The herbicidal composition according to claim 1, wherein said light-inducing herbicidal compound is one or more compounds selected from the substituted phenylpyrazole derivatives represented by the following general formula (I-1):

$$CI \longrightarrow X_1$$

$$X_2$$

$$X_1$$

$$X_2$$

$$(I-1)$$

$$R_1$$

wherein R represents

-Y1-R3

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wherein  $R^3$  represents a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkynyl group and  $Y^1$  represents -O- or -S-, -Y<sup>2</sup>CH( $R^4$ )CO-OR<sup>5</sup>

wherein R<sup>4</sup> represents a hydrogen atom or a  $(C_1-C_6)$  alkyl group, R<sup>5</sup> represents a hydrogen atom, a  $(C_1-C_6)$  alkyl group, a halo  $(C_1-C_6)$  alkyl group,

a (C<sub>2</sub>-C<sub>6</sub>) alkenyl group or a (C<sub>2</sub>-C<sub>6</sub>) alkynyl group, and Y<sup>2</sup> represents -O-, -S- or -NH-,

- COOCH(R4)CO-Y1-R5

wherein R4, R5 and Y1 are as defined above,

or -COOR6

wherein R<sup>6</sup> represents a  $(C_1-C_6)$  alkyl group, a  $(C_2-C_6)$  alkenyl group or a  $(C_2-C_6)$  alkynyl group;

R<sup>1</sup> represents a (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

R<sup>2</sup> represents a hydrogen atom, a (C<sub>1</sub>-C<sub>6</sub>) alkyl group or a halo (C<sub>1</sub>-C<sub>6</sub>) alkyl group;

X<sup>1</sup> and X<sup>2</sup> represent halogen atoms which may be the same or different;

Y represents -O-, -S-, -SO- or -SO<sub>2</sub>-;

and n represents an integer of 0 to 1.

3. The herbicidal composition according to claim 1, wherein said ethylenediamine alkoxylate is one or more compounds selected from the compounds represented by the following general formula (II):

$$H - (Po)_a - (Eo)_b$$
 $N - CH_2 - CH_2 - N$ 
 $(Eo)_f - (Po)_g - H$ 
 $H - (Po)_d - (Eo)_e$ 
 $(Eo)_h - (Po)_i - H$ 

wherein Eo represents -O- $CH_2CH_2$ -; Po represents -O- $CH(CH_3)CH_2$ -; and a, b, d, e, f, g, h and i represent integers of 1 to 20 which may be the same or different.

- 4. The herbicidal composition according to claim 1, wherein said ethylenediamine alkoxylate is an adduct of ethylenediamine and polyoxyethylene, an adduct of ethylenediamine and polyoxypropylene or an adduct of ethylenediamine and a polyoxyethylene-polyoxypropylene copolymer.
- 5. A herbicidal composition according to any one of claims 1 to 4, which contains one or more compounds selected from light-inducing herbicidal compounds in an amount of 0.01-10 parts by weight, an organophosphorus herbicidal compound in an amount of 1-60 parts by weight, an ethylenediamine alkoxylate in an amount of 0.1-25 parts by weight and an alcohol alkoxylate in an amount of 0.1-15 parts by weight, all per 100 parts by weight of the herbicidal composition.
- **6.** The herbicidal composition according to any one of claims 1 to 5, wherein said herbicidal composition is a suspension concentrate, a water dispersible granule or an aqueous preparation.
- 7. A method for using a herbicidal composition characterized by treating the objective weed or the soil with an effective amount of the herbicidal composition according to any one of claims 1 to 6 for the purpose of controlling the weeds harmful to crop plants.

## Patentansprüche

1. Herbizidzusammensetzung, enthaltend eine oder mehrere Verbindungen ausgewählt aus lichtinduzierenden Herbizidverbindungen und eine oder mehrere Verbindungen ausgewählt aus phosphororganischen Herbizidverbindungen als aktive Inhaltsstoffe, worin die lichtinduzierende Herbizidverbindung eine Verbindung ist, die durch die folgende allgemeine Formel (I) dargestellt wird:

worin P durch jedes von P1 bis P9 dargestellt wird:

$$CI \longrightarrow \begin{array}{c} X^1 \\ CI \longrightarrow \\ R^3 \end{array} \xrightarrow{P^1} \begin{array}{c} COOR^5 \\ P^2 \end{array} \xrightarrow{P^2} \begin{array}{c} X^1 \\ OR^6 \\ P^4 \end{array}$$

worin X<sup>1</sup>, Y, Y<sup>1</sup>, Y<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup>, R<sup>5</sup> und R<sup>6</sup> wie folgt definiert sind und Q eines von Q<sup>1</sup> bis Q<sup>11</sup> darstellt:

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R<sup>1</sup> eine (C<sub>1</sub>-C<sub>6</sub>)Alkylgruppe bedeutet;

R<sup>2</sup> ein Wasserstoffatom, eine (C<sub>1</sub>-C<sub>6</sub>)Alkylgruppe oder eine Halo(C<sub>1</sub>-C<sub>6</sub>)alkylgruppe bedeutet;

 $R^3$  eine  $(C_1-C_6)Alkylgruppe$ , eine  $Halo(C_1-C_6)Alkylgruppe$ , eine  $(C_3-C_6)Cycloalkylgruppe$ , eine  $(C_2-C_6)Alkenylgruppe$  oder eine  $(C_2-C_6)Alkinylgruppe$  bedeutet;

R<sup>4</sup> ein Wasserstoffatom oder eine (C<sub>1</sub>-C<sub>6</sub>)Alkyklgruppe bedeutet;

 $\mathsf{R}^5$  ein Wasserstoffatom, eine  $(\mathsf{C}_1\mathsf{-}\mathsf{C}_6)\mathsf{Alkylgruppe}$ , eine  $\mathsf{Halo}(\mathsf{C}_1\mathsf{-}\mathsf{C}_6)\mathsf{alkylgruppe}$ ,

eine (C2-C6)Alkenylgruppe oder eine (C2-C6)Alkinylgruppe bedeutet;

 $R^6$  eine  $(C_1-C_6)Alkylgruppe$ , eine  $(C_2-C_6)Alkenylgruppe$  oder eine  $(C_2-C_6)Alkinylgruppe$  bedeutet;

X¹ und X² Halogenatome, welche gleich oder verschieden sein können, bedeuten;

Y -O-, -S-, -SO- oder -SO<sub>2</sub>- bedeutet;

Y1 -O- oder -S- bedeutet;

Y<sup>2</sup> -O-, -S- oder -NH- bedeutet;

und n eine ganze Zahl von 0 bis 1 bedeutet,

und worin die phosphororganische Herbizidverbindung N-(Phosphonomethyl)glycerin oder ein Salz davon, 4-[Hydroxy(methyl)phosphino]-DL-homoalanin oder ein Salz davon oder 4-[Hydroxy(methyl)phosphino)-Lhomoalanyl-L-alanin oder ein Salz davon ist,

dadurch gekennzeichnet, dass die Herbizidzusammensetzung weiterhin ein Ethylendiaminalkoxylat und ein Alkoholalkoxylat als oberflächenaktive Verbindungen enthält,

worin das Alkoholalkoxylat eine Mischung aus einer oder mehreren

Verbindungen ausgewählt aus der Gruppe bestehend aus primärem Alkoholethoxylat, primärem Alkoholpropoxylat, primärem Alkoholethoxylat propoxylat, sekundärem Alkoholethoxylat propoxylat, sekundärem Alkoholethoxylat propoxylat, tertiärem Alkoholethoxylat, tertiärem Alkoholethoxylat und tertiärem Alkoholethoxylat propoxylat besteht.

2. Herbizidzusammensetzung gemäß Anspruch 1, worin die lichtinduzierende Herbizidverbindung eine oder mehrere Verbindungen ausgewählt aus den substituierten Phenylpyrazolderivaten, dargestellt durch die folgende allgemei-

ne Formel (I-1), ist:

 $Cl \qquad X_1 \qquad X_2 \qquad (I-1)$ 

worin R bedeutet:

15 -Y1-B3

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worin  $R^3$  eine  $(C_1-C_6)Alkylgruppe$ , eine  $Halo(C_1-C_6)Alkylgruppe$ , eine  $(C_2-C_6)Alkylgruppe$  oder eine  $(C_2-C_6)Alkylgruppe$  bedeutet und  $Y^1$ -O- oder -Sbedeutet,

-Y2CH(R4)CO-OR5

worin R<sup>4</sup> ein Wasserstoffatom oder eine  $(C_1-C_6)$ Alkylgruppe bedeutet, R<sup>5</sup> ein Wasserstoffatom, eine  $(C_1-C_6)$ Alkylgruppe, eine Halo $(C_1-C_6)$ Alkylgruppe, eine  $(C_2-C_6)$ Alkenylgruppe oder eine  $(C_2-C_6)$ Alkinylgruppe bedeutet und Y<sup>2</sup>-O-,

- S- oder -NH- bedeutet,

-COOCH(R4)CO-Y1-R5

worin R4, R5 und Y1 wie oben definiert sind,

25 oder -COOR<sup>6</sup>.

worin  $R^6$  eine  $(C_1-C_6)Alkylgruppe$ , eine  $(C_2-C_6)Alkenylgruppe$  oder eine  $(C_2-C_6)Alkylgruppe$  bedeutet;  $R^1$  eine  $(C_1-C_6)Alkylgruppe$  bedeutet;

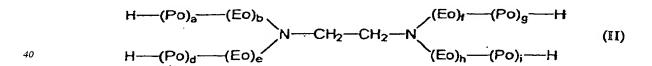
R<sup>2</sup> ein Wasserstoffatom, eine (C<sub>1</sub>-C<sub>6</sub>)Alkylgruppe oder eine Halo(C<sub>1</sub>-C<sub>6</sub>)alkylgruppe bedeutet;

X<sup>1</sup> und X<sup>2</sup> Halogenatome, welche gleich oder unterschiedlich sein können bedeuten;

Y -O-, -S-, -SO- oder -SO<sub>2</sub>- bedeutet;

und n eine ganze Zahl von 0 bis 1 bedeutet.

3. Herbizidzusammensetzung gemäß Anspruch 1, worin das Ethylendiaminalkoxylat eine oder mehrere Verbindungen ausgewählt aus den Verbindungen dargestellt durch die folgende allgemeine Formel (II) ist:



worin Eo -O-CH<sub>2</sub>CH<sub>2</sub>- bedeutet; Po -OCH(CH<sub>3</sub>)CH<sub>2</sub>- bedeutet; und a, b, d, e, f, g, h und i ganze Zahlen von 1 bis 20, welche gleich oder unterschiedlich sein können, bedeuten.

- 4. Herbizidzusammensetzung gemäß Anspruch 1, worin das Ethylendiaminalkoxylat ein Addukt aus Ethylendiamin und Polyoxyethylen, ein Addukt aus Ethylendiamin und Polyoxypropylen oder ein Addukt aus Ethylendiamin und einem Polyoxyethylen-Polyoxypropylen Copolymer ist.
- 50 5. Herbizidzusammensetzung gemäß einem der Ansprüche 1 bis 4, welche eine oder mehrere Verbindungen ausgewählt aus lichtinduzierenden Herbizidverbindungen in einer Menge von 0,01-10 Gewichtsteilen, eine phospororganische Herbizidverbindung in einer Menge von 1-60 Gewichtsteilen, ein Ethylendiaminalkoxylat in einer Menge von 0,1-25 Gewichtsteilen und ein Alkoholalkoxylat in einer Menge von 0,1-15 Gewichtsteilen enthält, alle bezogen auf 100 Gewichtsteile der Herbizidzusammensetzung.
  - 6. Herbizidzusammensetzung gemäß einem der Ansprüche 1 bis 5, worin die Herbizidzusammensetzung ein Suspensionskonzentrat, ein in Wasser dispergierbares Granulat oder eine wässrige Zubereitung ist.

7. Verfahren zur Verwendung einer Herbizidzusammensetzung, dadurch gekennzeichnet, dass das betreffende Unkraut oder die Erde mit einer wirksamen Menge der Herbizidzusammensetzung gemäß einem der Ansprüche 1 bis 6 behandelt wird zum Zweck der Kontrolle des Unkrauts, welches für Feldfrüchte schädlich ist.

### Revendications

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 Composition herbicide comprenant un ou plusieurs composés choisis parmi les composés herbicides photo-inducteurs et un ou plusieurs composés choisis parmi les composés herbicides organophosphorés en tant qu'ingrédients actifs de celle-ci, dans laquelle ledit composé herbicide photo-inducteur est un composé représenté par la formule générale suivante

dans laquelle ledit composé herbicide photo-inducteur est un composé représenté par la formule générale suivant (I) :

dans laquelle P représente l'un quelconque de P1 à P9 :

$$C \mapsto \begin{pmatrix} X^1 & X^1$$

dans lesquelles X1, Y, Y1, Y2, R3, R4, R5 et R6 sont tels que définis ci-dessous, et Q représente l'un quelconque de Q1 à Q11 :

$$-N \longrightarrow CF_3 \qquad N \longrightarrow CHF_2$$

$$Q^5 \qquad Q^6 \qquad Q^7$$

dans lesquelles

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R<sup>1</sup> représente un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) ;

R<sup>2</sup> représente un atome d'hydrogène, un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) ou un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) halogéné ;

 $R^3$  représente un groupe alkyle  $(C_1-C_6)$ , un groupe alkyle  $(C_1-C_6)$  halogéné, un groupe cycloalkyle  $(C_3-C_6)$ , un groupe alcényle  $(C_2-C_6)$  ou un groupe alcynyle  $(C_2-C_6)$ ;

R<sup>4</sup> représente un atome d'hydrogène ou un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) ;

 $R^5$  représente un atome d'hydrogène, un groupe alkyle  $(C_1-C_6)$ , un groupe alkyle  $(C_1-C_6)$  halogéné, un groupe alcényle  $(C_2-C_6)$  ou un groupe alcynyle  $(C_2-C_6)$ ;

 $R^6$  représente un groupe alkyle  $(C_1-C_6)$ , un groupe alcényle  $(C_2-C_6)$  ou un groupe alcynyle  $(C_2-C_6)$ ;

X1 et X2 représentent des atomes d'halogène pouvant être identiques ou différents ;

Y représente -O-, -S-, -SO- ou -SO<sub>2</sub>-;

Y1 représente -O- ou -S-;

Y<sup>2</sup> représente -O-, -S- ou -NH-;

et n représente un nombre entier 0 ou 1,

et dans laquelle ledit composé herbicide organophosphoré est la N- (phosphono-méthyl) glycine ou un sel de celleci, la 4-[hydroxy(méthyl)phosphino]-DL-homoalanine ou un sel de celle-ci ou la 4-[hydroxy(méthyl)-phosphino]-Lhomoalanyl-L-alanyl-L-alanine ou un sel de celle-ci,

caractérisé en ce que la composition herbicide contient en outre un alcoxylate d'éthylène diamine et un alcoxylate d'alcool en tant qu'agents tensio-actifs

dans laquelle ledit alcoxylate d'alcool est un mélange d'un ou plusieurs composés choisis parmi le groupe constitué de l'éthoxylate d'alcool primaire, du propoxylate d'alcool primaire, du propoxylate éthoxylate d'alcool primaire, de l'éthoxylate d'alcool secondaire, du propoxylate d'alcool secondaire, du propoxylate d'alcool tertiaire, du propoxylate d'alcool tertiaire.

2. Composition herbicide selon la revendication 1, dans laquelle ledit composé herbicide photo-inducteur est constitué d'un ou de plusieurs composés choisis parmi les dérivés de phénylpyrazole substitués représentés par la formule générale suivante (I-1):

$$CI \longrightarrow X_1$$
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dans laquelle R représente -Y1-R3

dans laquelle  $R^3$  représente un groupe alkyle ( $C_1$ - $C_6$ ), un groupe alkyle ( $C_1$ - $C_6$ ) halogéné, un groupe alcényle ( $C_2$ - $C_6$ ) ou un groupe alcynyle ( $C_2$ - $C_6$ ) et Y<sup>1</sup> représente -O- ou -S-, -Y<sup>1</sup>CH( $R^4$ )CO-OR<sup>5</sup>

dans laquelle R<sup>4</sup> représente un atome d'hydrogène ou un groupe alkyle  $(C_1-C_6)$ , R<sup>5</sup> représente un atome d'hydrogène, un groupe alkyle  $(C_1-C_6)$ , un groupe alkyle  $(C_1-C_6)$  halogéné, un groupe alcényle  $(C_2-C_6)$  ou un groupe alcynyle  $(C_2-C_6)$  et Y<sup>2</sup> représente -O-, -S- ou -NH-, -COOCH(R<sup>4</sup>)CO-Y<sup>1</sup>-R<sup>4</sup>

dans laquelle R<sup>4</sup>, R<sup>5</sup> et Y<sup>1</sup> sont tels que définis ci-dessus,

ou -COOR6

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dans laquelle  $R^6$  représente un groupe alkyle  $(C_1-C_6)$ , un groupe alcényle  $(C_2-C_6)$  ou un groupe alcynyle  $(C_2-C_6)$ ;

R<sup>1</sup> représente un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>);

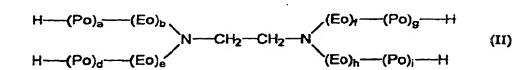
R<sup>2</sup> représente un atome d'hydrogène, un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) ou un groupe alkyle (C<sub>1</sub>-C<sub>6</sub>) halogéné ;

X1 et X2 représentent des atomes d'halogène pouvant être identiques ou différents ;

Y représente -O-, -S-, -SO- ou -SO<sub>2</sub>-;

et n représente un nombre entier 0 à 1.

3. Composition herbicide selon la revendication 1, dans laquelle ledit alcoxylate d'éthylènediamine est un ou plusieurs des composés choisis parmi les composés représentés par la formule générale suivante (II):



dans laquelle Eo représente -O-CH $_2$ CH $_2$ - ; Po représente -O-CH(CH $_3$ )CH $_2$ - ; et a, b, d, e, f, g, h et i représente des nombres entiers de 1 à 20 qui peuvent être identiques ou différents.

- 4. Composition herbicide selon la revendication 1, dans laquelle ledit alcoxylate d'éthylènediamine est un produit d'addition d'éthylènediamine et de polyoxyéthylène, un produit d'addition d'éthylènediamine et de polyoxyéthylène ou un produit d'addition d'éthylènediamine et d'un copolymère polyoxyéthylène-polyoxypropylène.
- 5. Composition herbicide selon l'une quelconque des revendications 1 à 4, qui contient un ou plusieurs composés choisis parmi les composés photo-induits en une quantité de 0,01 partie en poids à 10 parties en poids, un composé herbicide organophosphoré en une quantité de 1 partie en poids à 60 parties en poids, un alcoxylate d'éthylènediamine en une quantité de 0,1 partie en poids à 25 parties en poids et un alcoxylate d'alcool en une quantité de 0,1 partie en poids à 15 parties en poids, toutes quantités étant indiquées par rapport à 100 parties en poids de la composition herbicide.
- 6. Composition herbicide selon l'une quelconque des revendications 1 à 5, dans laquelle ladite composition herbicide est un concentré en suspension, un granulé hydrodispersible ou une préparation aqueuse.
  - 7. Procédé pour utiliser une composition herbicide caractérisé par le traitement des mauvaises herbes cibles ou du

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	sol grâce à une quantité suffisante de la composition herbicide selon l'une quelconque des revendications 1 à 6, dans le but de lutter contre les mauvaises herbes nuisibles aux cultures.
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